A CALIFORNIUM-252 FISSION SPECTRUM IRRADIATION FACILITY FOR NEUTRON REACTION RATE MEASUREMENTS

RADIOISOTOPES

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Spontaneous fission sources of ^{252}Cf, lightly encapsulated and with neutron source strengths approaching 10^{10} n/s, have been developed especially for integral cross-section measurements and neutron reaction rate calibrations. An irradiation facility at the National Bureau of Standards makes use of these sources in two well-investigated geometries. A free-field neutron flux in the range of 10^7 n/(cm^2 s) (10^5 n/mm$^2\cdot$s) and fluences of up to 10^{13} n/cm^2 (10^{11} n/mm^2) are established at the facility based only on a distance measurement and the absolute source strength of the national standard Ra-Be photoneutron source. The error in the ^{252}Cf source strength (±1.1%) dominates the total free-field flux uncertainty of ±1.4% (1σ). Neutron scattering effects in the source capsule and support structures and neutron return from concrete and earth boundaries have been calculated and investigated experimentally. In the worst case, they contribute ±0.7% to the total flux response uncertainty for all observed neutron reaction rates, including those with sensitivity to low-energy neutrons.

INTRODUCTION

Spontaneous fission sources of ^{252}Cf have been developed that are of substantial intensity and provide an unprecedented approximation to the ideal of an isolated point neutron source.[1] Fission neutrons at rates of $\sim 5 \times 10^9$ s^{-1} are emitted from these lightly encapsulated sources with an energy distribution studied more thoroughly than any other fast-neutron field.[2] When the source is placed in an isolated environment, a free-field fission neutron flux depending only on a source strength determination and a distance measurement can be established. Neutron fields of this type are being employed for cross-section measurements[3] and detector calibrations related to neutron standards in an irradiation facility developed specifically for this purpose at the National Bureau of Standards (NBS).

SOURCE DESCRIPTION

Each californium source is a disk-shaped deposit in an aluminum pellet encapsulated in a single stainless-steel cylinder. Assembly pieces for the source and an x-ray photograph of an assembled source capsule containing a 3-mg ^{252}Cf deposit are shown in Fig. 1. Sintered particles of $Cf_2O_2SO_4$ are dropped into the aluminum pellet cylinder, and the aluminum powder is pressed in to fill a central tapered bore. The pellet is placed in the steel capsule, and the cap is welded in place for closure. The threaded knob is for attaching a source-handling wand. Physical specifications for the source components are given in Table I. The position of the californium deposit relative to capsule surfaces is known to ±0.5 mm. This estimate is based on constraints of fabrication and has been verified by means of x-ray photographs. Neutron emission due to (α,n) reactions in either oxygen or aluminum is negligible. An upper limit of $\sim 2 \times 10^{-6}$ for the ratio of (α, n) to fission neutrons is estimated for thorough mixing of californium with aluminum or oxygen.

Inelastic neutron scatter in the source capsule is the primary perturbing effect for the free-field

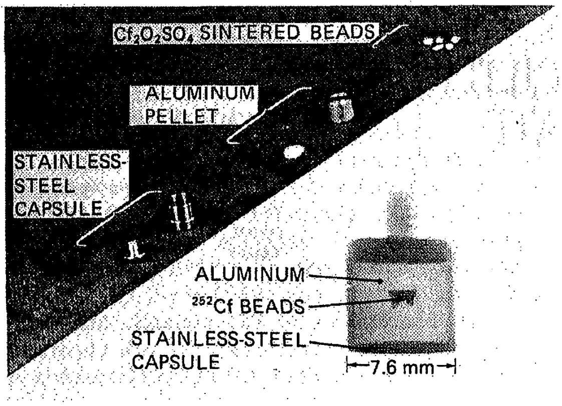

Fig. 1. Californium-252 source assembly components and x ray of assembled 3-mg source.

TABLE I

Physical Specifications for the Source Components

	Mass	Thickness of Cylinder Wall (mm)
Sintered beads ($Cf_2O_2SO_4$): (localized displacement ~1.4 mm^3)	~3 mg ^{252}Cf	
Aluminum pellet: (6.4 mm diam × 5.7 mm long)	0.48 g	2.16 ± 0.1
Stainless-steel capsule: (7.6 mm diam × 7.6 mm long; Type 304 stainless steel)	1.39 g	0.53 + 0.03
Capsule total:	1.87 g	2.7

fission spectrum flux. The fraction of the neutrons that undergo one inelastic scatter in the aluminum or steel of the capsule is (0.7 ± 0.3)%. Elastic scattering is of concern insofar as the emerging neutrons are rendered anisotropic since the source strength is determined by a method of total neutron absorption in 4π solid angle, while the detector response is commonly measured at specific angles. This anisotropy has been investigated with fission chambers set on opposite sides of the source. Responses in this arrangement for source rotations about the cylinder axis of the source capsule and for rotations perpendicular to that axis show an anisotropic emission. Based on these rotation measurements, the ratio of 4π emission to emission in a direction perpendicular to the cylinder axis (the normal source-detector arrangement) is found to be 0.992 ± 0.003.

Neutron source strengths for five sources presently available range from 0.7 to 5×10^9 n/s. These source strengths are determined in the NBS Manganous Sulfate Bath Facility against the internationally compared standard Ra-Be photoneutron source, NBS-I. The error in the ^{252}Cf source strengths is ±1.1% (1σ), with most of the error coming from the source strength calibrations of NBS-I.

THE ^{252}Cf FISSION NEUTRON SPECTRUM

The energy distribution of fission neutrons has been widely studied both experimentally and theoretically. A recent evaluation of eight documented spectrometry measurements for ^{252}Cf indicates substantial agreement among observed spectra over the bulk of the energy range.[2,4] The one-parameter Maxwellian function $\chi(E) = 0.663\sqrt{E} \times \exp(-1.5\,E/2.13)$ with empirical adjustments of $<2\%$ fits the compiled data well over the energy range from 0.25 to 8 MeV. Somewhat larger adjustments of the Maxwellian function outside this energy range are required to give a least-squares fit of the data over all energies. Spectrum uncertainties estimated in the evaluation were based on the departure of experimental data subsets from the adjusted Maxwellian. An eight-group tabulation of the evaluated ^{252}Cf fission spectrum including uncertainties is given in Table II, along with results for the ^{235}U thermal-induced fission spectrum, which was included in the same evaluation. Spectrum uncertainties, given at both the 67 and 95% confidence levels, make it possible to assign an error to a computed fission-spectrum-averaged cross section when comparisons with experiment are being investigated. The simultaneous evaluation of both these fission spectra also lends confidence to the application of detector calibrations with ^{252}Cf to reactor neutron environments driven by ^{235}U fission neutrons.

NEUTRON FLUX SPECIFICATION AND IRRADIATION FACILITIES

Two irradiation facilities are available at NBS. An isolated, lightweight source-detector assembly in a room with an open ceiling is employed for measurements that are not selectively sensitive to low-energy neutrons. For minimal boundary return of neutrons, an alternative arrangement employing an outdoor mast places the same source-detector assembly 5 m above the earth. The source-detector assembly is shown in Fig. 2

TABLE II

Eight-Group Tabulation of Fission Neutron Spectrum Evaluation with Uncertainties

Energy Boundaries	Californium-252 (Spontaneous Fission)			Uranium-235 (Thermal-Neutron-Induced Fission)		
	Group Flux	Error		Group Flux	Error	
		1σ (%)	2σ (%)		1σ (%)	2σ (%)
0.0 – 0.25	0.047	±13	±26	0.054	±16	±32
0.25 – 0.8	0.184	±1.1	±3.3	0.197	±4.1	±6.2
0.8 – 1.5	0.220	±1.8	±3.6	0.229	±3.0	±4.8
1.5 – 2.3	0.194	±1.0	±3.1	0.195	±3.1	±5.2
2.3 – 3.7	0.200	±2.0	±3.0	0.192	±2.0	±3.0
3.7 – 8	0.146	±2.1	±4.8	0.127	±4.8	±8.0
8 – 12	0.0087	±8.5	±17	0.0056	±5.3	±11
12 – 20	(0.00058)			(0.00026)		

along with distances to boundaries in the two facilities that give rise to a neutron return flux. Neutron field parameters for a nominal 5-cm (50-mm) source-to-detector distance excluding neutron return from the environment are given in Table III. The source strength error dominates. Further efforts are required, therefore, including international participation, if reducing traditional absolute neutron source strength errors to below ±1% is desired. Compensated flux geometry refers to our usual experimental practice of placing nearly identical detectors on opposite sides of, and equidistant from, the source. The first-order distance error is then associated with the separation of detectors; the uncertainty in the position of the source deposit becomes second order. In addition, the source may be rotated about its cylinder axis during irradiations to further ensure proper spatial averaging of the neutron flux at the detectors.

Neutron return from the environment including irradiation support structures, and the resultant backgrounds for important classes of neutron detectors, are given in Table IV for a 5-cm (50-mm) source-to-detector distance. The reaction rate perturbations for fission detectors are estimated from response-versus-distance measurements undertaken in the indoor room and outdoors where the return is exclusively from the plane of the earth. Cadmium ratios of ~9 are observed for $^{235}U(n,f)$ in the boundary return flux. Environmental return flux backgrounds above 0.4 eV are estimated from computation:

TABLE III

Neutron Field Parameters

Free-field fission neutron flux	1×10^7 n/(cm^2 s) (1×10^5 n/mm$^2 \cdot$ s)
Source decay rate	2.3%/month
Free-field fluence for 100-h exposure	4×10^{12} n/cm^2 (4×10^{10} n/mm^2)
Source capsule scattering (inelastic plus net elastic inscatter)	1.1%
Gamma-ray exposure (2.8 yr after separation)	~300 R/h
Error components for free-field fission neutron flux (1σ):	
source strength	±1.1%
source capsule and support scattering	±0.7% (max)
distance measurements (typical for compensated flux geometry)	±0.6%
Total free-field flux error (root-mean-square sum):	±1.4% (1σ)

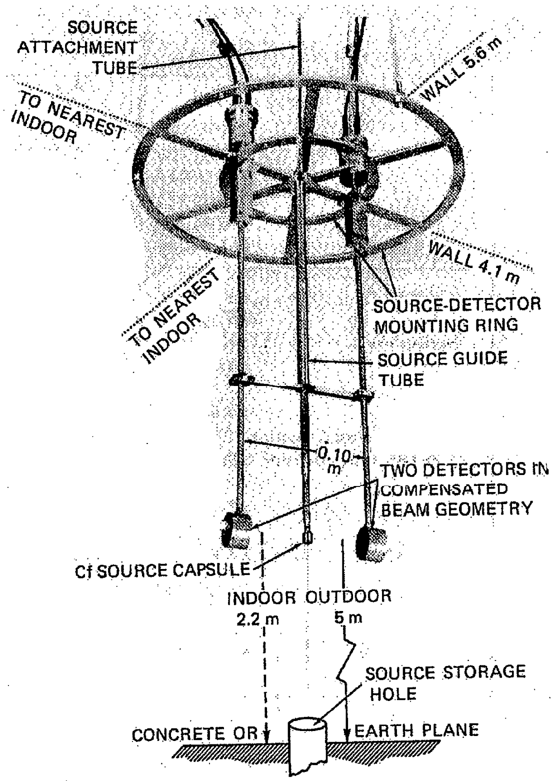

Fig. 2. Source detector arrangement for indoor and outdoor irradiation facility. Detector placement is undertaken with source in storage hole. Source is withdrawn to position shown for irradiation.

1. Albedo from boundaries of the indoor room is based on a discrete ordinates calculation of a spherical cavity in concrete.
2. The effect of support structures excluding detector holders is due almost entirely to the aluminum source support tubes and is estimated on the basis of single scattering events in individual support pieces.
3. Air scatter estimates are based on published analytical formulations involving point sources and a simple scattering kernel.[5]

The estimate of albedo from the ground for the outdoor mast was based on calculations of neutron reflection from an infinite plane.

MEASUREMENTS WITH ^{252}Cf FISSION SPECTRUM NEUTRONS

Isolated ^{252}Cf fission neutron sources have been applied to cross-section measurements and re-

TABLE IV

Neutron Return from the Environment Including Irradiation Support Structures and the Resulting Backgrounds for Important Classes of Neutron Detectors for a 5-cm (50-mm) Source-to-Detector Distance

	Indoor Room	Outdoor Mast
Distance to nearest boundary	2.3 m	5 m
Environmental return flux background above 0.4 eV		
Albedo from boundaries	$\lesssim$0.07%	<0.004%
Irradiation support structures	0.3%	0.3%
Air scatter	<0.1%	<0.1%
Net reaction rate perturbation due to scattered neutrons (source capsule included)		
^{235}U(n,f) detector with cadmium enclosure	(1.4 ± 0.6)%	(1.2 ± 0.6)%
^{238}U(n,f) and higher threshold detectors	(0.4 ± 0.7)%	(0.4 ± 0.7)%
$1/v$ detector with cadmium enclosure	~2½%	

lated calibrations at various laboratories in recent years.[6-8] At NBS, irradiations to a specified fission neutron fluence have provided calibrations for specialized health physics monitors and for the determination of flux transfer factors for ^{239}Pu(n,f) and ^{32}S(n,p) detectors used to monitor neutron fluxes in and around fast burst reactors. Special high-fluence irradiations also have been undertaken for the study of radiation damage in transistors.

Fission cross-section measurements with ^{252}Cf fission neutrons have been carried out with double fission chambers and the NBS set of reference and working fissionable deposits.[3,9] Recently completed fissionable deposit isotopic mass intercomparisons and comparisons of observed and calculated anisotropy of source capsule transmission provide an updated experimental result for the fission cross section of ^{235}U:

$$\bar{\sigma}_f\,(^{235}\mathrm{U},\chi_{\mathrm{Cf}}) = 1205 \pm 27 \text{ mb}$$

This value falls below that calculated with the ^{235}U(n,f) energy-dependent cross sections of ENDF/B-IV by 3.0%. The error in this cross-section validation measurement attributable to ^{252}Cf fission spectrum uncertainties is less than ±0.2%.

Calibration irradiations in the BIG-TEN Critical Assembly at Los Alamos Scientific Laboratory, a benchmark for breeder reactor fuels and materials dosimetry, have been monitored with the NBS double fission chambers.[10] One component of these measurements was to establish a neutron flux level in such an environment by means of a direct flux transfer from the NBS ^{252}Cf fission neutron field. The transfer to the central BIG-TEN field, accomplished with a ^{239}Pu-loaded fission chamber, does not depend significantly on any neutron reaction cross section. Since the ^{239}Pu fission cross section is largely independent of energy for both spectra, the spectrum correction for the flux transfer, $\bar{\sigma}_f(^{239}\text{Pu},\chi_{Cf})/\bar{\sigma}_f(^{239}\text{Pu}, \text{BIG-TEN})$, is close to unity; the calculated value[9,10] is 1.11 ± 0.015. The neutron flux level for BIG-TEN irradiations of high-power activation dosimeters was established at 8.2×10^{10} n/(cm^2 s) (8.2×10^8 n/mm$^2 \cdot$ s), with a total error, exclusive of cavity perturbations, of less than $\pm 2\frac{1}{2}\%$ (1σ).

An absolute spectrum-averaged cross section for $^{238}\text{U}(n,f)$ can be obtained on the basis of this flux transfer measurement. For the center of BIG-TEN, the cross section determined with a ^{238}U fission chamber containing a natural uranium deposit of known mass is

$$\bar{\sigma}_f(^{238}\text{U, BIG-TEN}) = 50.7 \pm 1.5 \text{ mb}$$

The computed value for the BIG-TEN spectrum using the ENDF/B-IV cross sections is 46 mb. The discrepancy of 10% between measured and computed values is substantial and somewhat greater than expected for similar fast critical assemblies.[11]

CONCLUSION

Fission neutrons from the spontaneous fission of ^{252}Cf are available at flux levels of $\gtrsim 10^7$, which compare favorably with zero-power reactor environments. As a measurement base for establishing fast-neutron reaction rates, they are unmatched with regard to both spectrum definition and critically evaluated accuracy of fission neutron flux. However, to exploit the maximum potential for purposes of calibration and nuclear data validation, the irradiation facility must be well studied. Small neutron return fluxes can be important for certain types of detectors, and scattering effects as well as source-detector geometry must be carefully investigated. For the facility described here, the uncertainty in these experimental problems has been reduced to below $\pm 1\%$ (1σ). The dominating error then is the determination of the absolute neutron source strength presently set at $\pm 1.1\%$ (1σ). The total error for a free-field fission neutron fluence at the NBS ^{252}Cf irradiation facility is $\pm 1.4\%$ (1σ).

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